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Single channel waveguide with roughness

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We recently introduced a concept of a neutron waveguide for enhancing the intensity of the evanescent wave inside the sample next to the waveguide. This allows for studying tribology mechanisms of soft matter/solid interfaces using grazing incidence neutron spin echo spectroscopy (GINSES) and small angle neutron scattering (GISANS). In several experiments, the number of layer structures in the waveguide was chosen to be 1 and 3. Both waveguides increased the scattering intensity of GISANS (and GINSES) experiments successfully, while the simpler waveguide produced additional small angle scattering that was not rationalized so far. The roughness on the 100nm and μm length scale may produce additional background at smaller angles. Comparisons are drawn using the Distorted Wave Born Approximation (DWBA), which is especially useful for the roughness discussion.

KEYWORDS: Neutron Resonator, Neutron Waveguide, Distorted Wave Born Approximation, Roughness, Grazing Incidence Small Angle Neutron Scattering

1. Introduction

The evanescent wave is a tunneling wave that develops in a grazing incidence small angle neutron scattering experiment when staying with the incidence angle below the critical angle of total reflection. Then, classically, all neutrons are reflected and only from the near surface waves neutrons can be scattered by the sample. The exponential decay length (also called scattering depth Λ) provides the possibility of depth profiling. Thus, conclusions can be drawn about the near surface structure [1] and dynamics [2], which is highly important for understanding tribological mechanisms.

The spectroscopic neutron experiments (GINSES) demand for highest impinging intensities because the scattering volumes are small and because of the detailed analysis of small energy changes in general. The recently introduced neutron waveguides [3, 4] provide an enhancement of the evanescent wave by factors of ca. 3-7 [4] from which increased scattering intensities emerge. A waveguide allows for constructive interference of multiple reflections inside the guide, the intensity of which can extend into the exterior sample. We argued that the unwanted scattering of the resonator structure is reduced by a lower number of layers in the waveguide. The surface roughness, and likely the waviness at length scales of μm reduces the constructive interference and therefore inhibits highest gains in the actual grazing incidence scattering experiment. A specially polished silicon slab was produced by the company Zeiss.

These findings are highly plausible, while there was unwanted small angle scattering observed for the simpler resonator with less layers. This effect is observed in experiments and could be reproduced in DWBA simulations. In this manuscript we present the findings and rationalize the results with a simple theory. For further experiments we finally would suggest the 3-fold layer structure of the resonator.

2. The observation of the simplest resonator

The simplest resonator consists of Ti/Pt/Ti layers with thicknesses of 130/320/130Å on top of a large silicon slab with dimensions of $150 \times 80 \times 40$ mm^3. The slab was polished to the smallest possible waviness with wavelengths of 1-10mm with amplitudes of ca. 1nm by the company Zeiss. All materials are non-magnetic such that they ideally serve for neutron spin-echo experiments without destroying the neutron polarization. The selected sample was a microemulsion with film contrast (D_2O, d-decane, and hydrogenous $C_{10}E_4$ surfactant at volume fractions of 41.5/41.5/17). The scattering obtained from GISANS measurements on MARIA [5] of the microemulsion is shown in Figure 1. The instrument was operated at a wavelength of 10 Å with $\delta\lambda/\lambda = 0.1$ in standard configuration with a beam divergence of 0.02°. The beam penetrates the silicon slab, impinges the resonator, and the evanescent wave behind is in the sample. From previous experiments [1] we know that 4 perfect lamellar domains of water or oil are found adjacent to the solid-liquid interface before the order decays over perforated lamellae to the bicontinuous structure. In film contrast, only the surfactant film of approx. 10 to 12Å thickness is visible with the oil/water domains having a thickness in the range of approx. 100 to 110Å. The scattering length densities for Si/Ti/Pt/D_2O/d-decane/surfactant are 2.07/-1.95/6.36/ 6.36/6.49/0.12 $\times 10^{-6}$Å^{-2}. The specular reflex is found at approx. $Q_z = 0.007$Å^{-1}, while we define $Q_z = 0$Å^{-1} to be at the primary intensity. The scattering is elevated at the Bragg peak positions $Q_z \approx 0.04$ (first order) and 0.08Å^{-1} (second order) by a factor of 3 to 7 when subtracting the background (Fig. 1). A small angle scattering contribution appears at $Q_z < 0.03$Å^{-1} which we attribute to a diffuse scattered background from the resonator and the osmotic compressibility of the microemulsion. In section 3 we show with simulations that such a contribution is easily obtained from surface roughness scattering. An increase in scattering from the microemulsion osmotic compressibility would result in an increase of the first order Bragg peak at $Q_z \approx 0.04$ Å^{-1} but with a flat intensity at lower Q_z. The overall diffuse signal is rather high (in units of Fig. 1 of approx. 30) and superimposes to the first

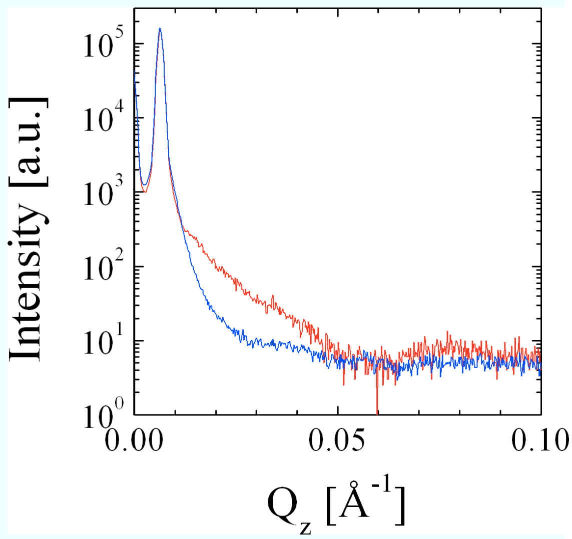

Fig. 1. GISANS experiments on a film-contrast microemulsion with a simple resonator (red) and with a simple silicon slab (blue) [4]. The specular reflex is found along the normal direction at $Q_z \approx 0.007$Å^{-1}, the microemulsion peaks at ca. 0.04 and 0.08Å^{-1}. The resonator seems to cause additional small angle scattering at $Q_z < 0.03$Å^{-1}.

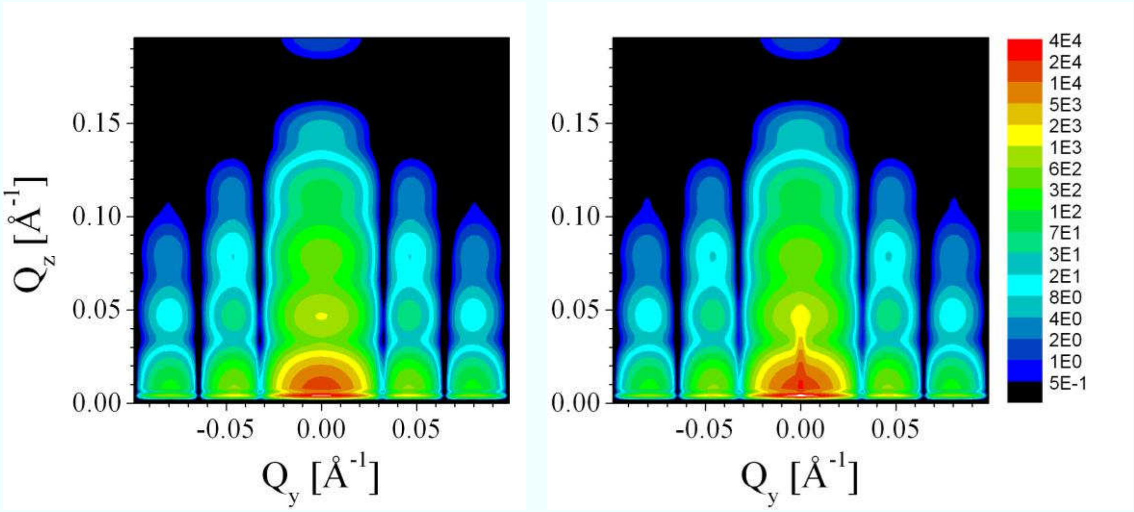

Fig. 2. DWBA simulations of a GISANS experiment with a simple resonator without (left) and with (right) roughness of the sputtered layers. The colors are displayed on a logarithmic scale.

order Bragg peak of the microemulsion at Q_z = 0.04Å^{-1}. So the gain factor might be smaller than 7 that initially was optimistically estimated. In a reflectivity scan, the unwanted resonator background of the third order Bragg peak at Q_z = 0.03Å^{-1} (not observed in Fig. 1, rationalized by the resonator thickness of 130+320+130Å, i.e. Q_{Bragg} = 0.01Å^{-1}) was rather low in comparison to a threefold resonator structure, but this ratio reversed at even higher Q_z [4]. In any case, we see the strong influence of the resonator background when the length scales of the resonator (580Å) and the sample (110Å) are comparable. So the advantage of less layers in the resonator for reduced unwanted background in GISANS and reflectivity measurements needs to be reconsidered. The additional small angle scattering is inacceptable, and the high-Q_z performance should also be at the highest level for GINSES measurements.

3. DWBA Simulations

The following model was developed using the BornAgain software [6] (see Fig. 2): The microemulsion was modeled as 100×100Å^2 patches of water, surfactant, oil, surfactant, etc. (total of 2 repetitions) with thicknesses of 100, 10, 100, and 10Å along the z-axis. These patches were correlated in the x-y-plane using a paracrystal model. The beam in the simulation was "ideal" (no divergence) since we want to focus only on the effect of the roughness with this simulation. While the microemulsion film-contrast generates a Bragg peak at around Q_z = 0.04Å^{-1}, the artificial paracrystal model generates Bragg peaks along the Q_y (and Q_x) axis. The "repetition" is artificial and should not be compared to the experiment while the central scattering along the Q_z axis is rather representative. When we compare an ideally plain structure of the resonator, no indication of superimposed scattering from the resonator is visible. Contrarily, when assuming finite correlated roughness (that results from the silicon polishing), a parasitic small angle signal is superimposed on the microemulsion signal, which is visible at Q_y=0 in Figure 2 and in the line cuts along the Q_z axis (Fig. 3). Besides the Yoneda scattering at very low angles at 0.003Å^{-1}, both systems show many fringes with a repeat distance of ca. 110Å resulting from the microemulsion under film contrast. Including surface roughness and keeping all other parameters of the simulation the same, a diffuse (additive) background appears at ca. Q_z < 0.03Å^{-1}. The roughness introduced here has a rms of 1nm, a correlation length of

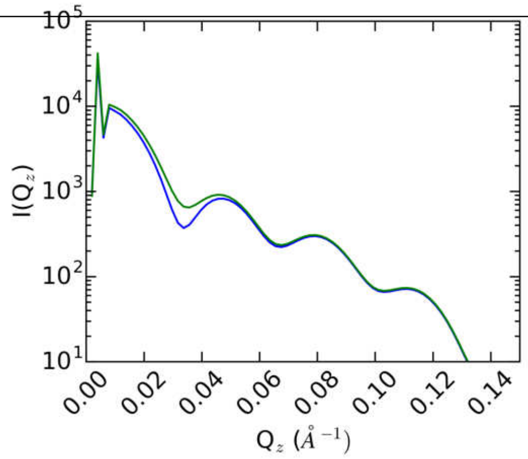

Fig. 3. Line-cut along the Q_z axis for the examples of Fig. 2. The blue line with the lower intensity at $Q_z = 0.03Å^{-1}$, shows the line cut of the ideal resonator, the green line shows the additional scattering due to the surface roughness of the resonator.

50nm and a Hurst parameter of 0.5. Astonishingly, no scattering from the resonator is superimposed, but this is a feature of the DWBA approximation: The layers of the resonator were modeled as distinct layers where refraction occurs, and so the scattering of these structures does not appear in the model. In the present example, the modeling of the diffuse scattering from the layer roughness seems to be sufficient. When comparing the magnitude of the diffuse scattering with the experiments, the amplitude of the surface roughness (taken from the manufacturers specifications) seems to be too low. The general parameterization of roughness in a precise manner is not trivial and requires a considerable experimental effort. The simulations suggest that the true amplitude is rather ~30Å instead of 10Å amplitude (when comparing simulations with the experiment). We would like to stress that the gross decay of intensity in Fig. 3 goes with Q_z^2 according to a single waveguide structure. Reflectometry measurements can provide an excellent insight into the details of the surface roughness. Work is ongoing with the resonator without microemulsion sample in order to better understand the experimentally observed diffuse GISANS scattering.

4. Scattering of a 3-fold resonator

The first resonator that has been built was a threefold resonator with Ti/Pt/Ti/Pt/Ti/Pt/Ti layers with thicknesses of 130/320/130/320/130/320/130Å. It proved to be highly useful for GINSES experiments on lipid bilayer dynamics at the solid/liquid interface [7]. When we look on the GISANS experiment (Fig. 4) of this system, the diffuse scattering from roughness is less important, and the scattering of the resonator itself is visible. There are hints that the DWBA also predicts such Bragg peaks as the one coming from the neutron waveguide structure at Q_z=0.02Å^{-1} when there is finite surface roughness. Again, the parameterization of the roughness is a nontrivial task and the origin has to be further investigated, since it is one of the crucial points of the resonator performance. Best possible polishing lead to a very low roughness, but variations with rather long correlation length could still have a significant influence on the resonator performance. We would like to stress that the gross dependence of the reflectivity curve in Ref. 7, Fig.7 goes with Q_z^4 for the 3-fold resonator having a volume property contrarily to the single resonator structure, indicating a better performance of the 3-fold resonator in terms of background at higher Q_z.

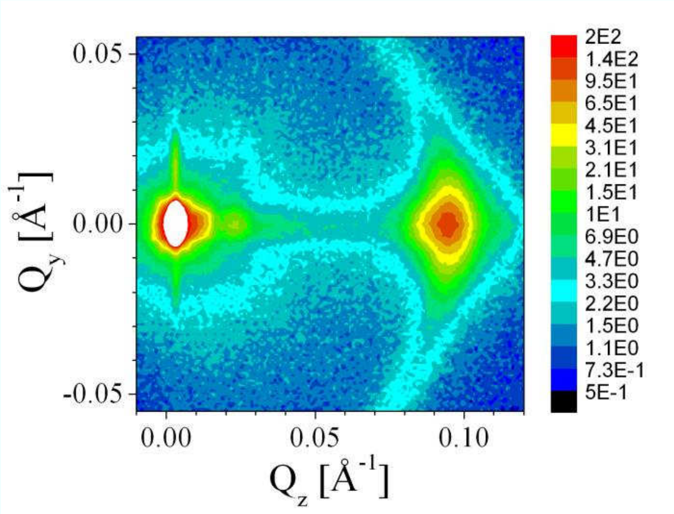

Fig. 4. GISANS experiments with the threefold resonator and a lipid double layer stack (in D_2O) on top of the solid. The Bragg peak of the lipid bilayers is at ca. Q_z = 0.095Å^{-1}, while the resonator Bragg peak is found at ca. Q_z = 0.02Å^{-1}.

5. Conclusion

We have seen that surface roughness plays an important role in GISANS experiments to describe a parasitic background in terms of small angle scattering when the diffuse scattering dominates, and in terms of Bragg peaks when the resonator consists of more layers (3-fold structure) than in the simplest case (1-fold structure). The gain factors have not been determined to a sufficient degree of precision due to that background in the case of the simple resonator. Strategies to achieve intensity gains for GISANS and GINSES experiments larger than the experimentally confirmed value of ca. 3 need to be developed. Simulations with the BornAgain software proved to be a valuable tool to understand the contributions of resonator and sample in grazing incidence neutron scattering experiments. The influence of surface roughness and parastitic scattering has to be minimized, where an improved polishing of the resonator substrate alone does not seem to lead to the desired low surface roughness in the end. Further investigations of the neutron resonators will help improving the gain factor of this technique.

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